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Symmetrized-Master-Equation Approach to Spontaneous Emission.

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Summary. — Spontaneous emission of radiation from extended systems of N atoms (or molecules) is studied by using the master-equation approach. Two internal states are involved for each atom considered as spin $\frac{1}{2}$. In order to treat the master equation, we use the properties of the irreducible representations of the group of permutations over the spin variables. Assuming that the atoms have been equivalently excited, a relevant part of the atomic density operator $\varrho(t)$ can be defined. This relevant part of $\varrho(t)$ is invariant under any permutation over the spin variables. It obeys a tractable equation of motion that we will call the « symmetrized » master equation or S.M.E. This equation is an interpolation between the master equation for an incoherent decay and the one associated to the co-operative decay in small samples. The preparation of the atomic system is also discussed. The S.M.E. will be valid for atomic systems prepared either by an incoherent excitation process or by intense light pulses which leave the system in highly excited states. Therefore our model will not be used when the atoms have been prepared near the superradiant state. Approximate solutions of S.M.E. are given in the limit of large N . It is found that co-operative effects appear for a larger concentration than that predicted by previous works. This disagreement results from the fact that in the S.M.E. the atomic co-operation number is not *a priori* constant. For large concentrations, the incoherent process, which still prevails at the beginning of emission, brings about a retardation of co-operative emission. During the fast decay of atoms, the behaviour of systems described in S.M.E. is found to be identical to the semi-classical one. Effects of inhomogeneous broadening are also considered. In this case, the incoherent process quenches the co-operative emission except for very large concentrations. Finally, detection aspects of the radiation field are described for detectors set far from the source.

1. — Introduction.

Since the classic paper of DICKE ⁽¹⁾, the co-operative emission of radiation by a collection of N two-level atoms (or molecules) has been extensively studied ^(2,4). Physically, these studies can be divided in two groups. In the first one, the atoms are contained in a pencil-shaped cavity and coupled to the resonant axial mode ⁽²⁾. The escape of emitted photons from the cavity is phenomenologically described by a cavity loss mechanism ^(2c). As all atoms in the cavity are equivalent with respect to the field, the co-operative effects will be maximum. Then, if the system has been coherently excited, it will emit an intense pulse of light. In the other case ^(3,4), the atoms are assumed to interact with the entire electromagnetic field, which is initially in the vacuum state. This is the case of spontaneous emission. As the interactions between pairs of atoms via the electromagnetic field are strongly dependent on the interatomic distances, the co-operative effects, *i.e.* the enhancement of the intensity of radiation and the directivity of emission, will be very sensitive to the size, the shape and the concentration of samples. For samples whose linear dimensions are much smaller than the radiation wavelength and without inhomogeneous broadening, the atoms are indistinguishable one from each other ^(3,4). Then, a pulse with an intensity proportional to N^2 can take place, but without directivity. However in the optical frequency range, such small samples are not realistic.

In this paper we are interested in the spontaneous emission of large samples. The atoms + field Hamiltonian is

$$H = H_0 + V,$$

where

$$(1.1) \quad \left\{ \begin{array}{l} H_0 = \sum_j \omega_j R_{3j} + \sum_k \omega_k a_k^\dagger a_k, \\ V = \sum_{k,j} (\alpha_{kj} \exp[i\mathbf{k} \cdot \mathbf{r}_j] a_k R_j^\dagger + \text{h.c.}) \end{array} \right.$$

⁽¹⁾ R. H. DICKE: *Phys. Rev.*, **93**, 99 (1954).

⁽²⁾ a) R. BONIFACIO and G. PREPARATA: *Lett. Nuovo Cimento*, **1**, 887 (1969); *Phys. Rev. A*, **2**, 336 (1970); b) G. SCHARF: *Helv. Phys. Acta*, **43**, 806 (1970); c) R. BONIFACIO, P. SCHWENDIMANN and F. HAAKE: *Phys. Rev. A*, **4**, 302, 854 (1971); d) V. DEGIORGIO and F. GHIEMMETTI: *Phys. Rev. A*, **4**, 2415 (1971); V. DEGIORGIO: *Opt. Comm.*, **2**, 236 (1971).

⁽³⁾ a) V. ERNST and P. STEHLE: *Phys. Rev.*, **176**, 1456 (1968); b) R. H. LEHMBERG: *Phys. Rev.*, **181**, 32 (1969); c) D. DIALETIS: *Phys. Rev. A*, **2**, 599 (1970); d) N. E. REHLER and J. H. EBERLY: *Phys. Rev. A*, **3**, 1735 (1971).

⁽⁴⁾ a) G. S. AGARWAL: *Phys. Rev. A*, **2**, 2038 (1970); b) G. S. AGARWAL: *Phys. Rev. A*, **4**, 1791 (1971).



and

$$\alpha_{kj} = -i\omega_j \sqrt{\frac{2\pi}{L^3 \hbar \omega_k}} \boldsymbol{\mu}_j \cdot \boldsymbol{\epsilon}_k,$$

ω_j is the frequency transition between upper and lower levels of atom j , $\mathbf{r}_j$ its position in the sample. $R_j^\pm$, R_{3j} are equivalent to the Pauli spin operators.

In this case, the atoms are generally not equivalent. Mathematically, it implies that the square length of the collective angular-momentum operator $\mathbf{R}$ does not commute with H ⁽⁵⁾. The co-operation number of the atomic system which is defined with the help of the expectation value of R^2 will not remain constant during the decay process. The variation of the co-operation number implies the mixing between different eigenstates of R^2 , or in other words leads to a partially incoherent emission process.

Hitherto the authors who have studied the co-operative emission of radiation in large samples ^(3a,3d) or in samples presenting an inhomogeneous broadening ^(4b) have neglected this property which can strongly modify the behaviour of the system. The purpose of this paper is to develop a tractable model taking the nonconservation of the square length of the angular momentum into account.

Often, a master-equation approach is used to study the interaction of a system of discrete levels with a continuum ⁽⁶⁾. For spontaneous emission of two-level atoms, a master equation for the reduced density operator of the atomic system has been derived within the second-order Born and Markoff approximations ^(3b,4a). These approximations are discussed in Appendix A with respect to the present problem. The Born-Markoff approximations imply in the case of N decaying atoms that retardation effects due to the propagation of light between atoms are neglected. At time $t=0$ each atom begins to decay by reacting with its own radiation field. The interaction between an atom and the radiation field emitted by the other ones will take place only at time $t \simeq L/C$, where L is the length of the sample. So, if this delay is much smaller than the radiative lifetime of a single atom Γ^{-1} , the co-operative effects will be considered to start at $t=0$ ^(3a,7,8). Then the Born-Markoff approximations are valid only if

$$(1.2) \quad \frac{L}{C} \ll \Gamma^{-1}.$$

We show in Appendix A that the Born approximation is not an improvement to the Born-Markoff approximations. Indeed, the Born approximation intro-

⁽⁵⁾ Or, more generally, the square length of any collective angular-momentum operator $\mathbf{R}_k = \sum_j \mathbf{R}_j \exp[i\mathbf{k} \cdot \mathbf{r}_j]$ does not commute with H .

⁽⁶⁾ P. N. ARGYRES and P. L. KELLEY: *Phys. Rev.*, **134**, A 98 (1964).

⁽⁷⁾ J. P. BARRAT: *Journ. Phys. Rad.*, **20**, 541 (1959).

⁽⁸⁾ E. RESSAYRE and A. TALLET: *Lett. Nuovo Cimento*, **5**, 1105 (1972).

duces a delay in the interaction between any pair of atoms which is twice the exact delay. Then for intermediate cases ($L/C \simeq \Gamma^{-1}$) neither the Born approximation nor the Born-Markoff approximations would be valid.

Afterwards we will assume that the condition (1.2) is met. Then the spontaneous emission of radiation by an extended atomic system can be described by the master equation derived within the Born and Markoff approximations. The Liouvillian operator associated to the equation of motion for the reduced density operator $\varrho(t)$ depends explicitly on the positions of atoms in the sample so that the master equation cannot be exactly solved whatever the choice of the atomic basis. For the treatment of the master equation, we choose the orthonormal basis of collective spin states and we use the properties of this basis which are related to the properties of the group of permutations over the spin variables. Then, we develop a model which is valid within an accuracy of order $1/N$, when the atoms are assumed equivalently excited. We obtain a tractable equation of motion for the part of $\varrho(t)$ which gives the main contribution to the decay process. The mathematical treatment (Sect. 2) is performed for atomic systems without inhomogeneous broadening. In these conditions, the equation of motion for the relevant part of $\varrho(t)$ depends on a parameter β , symmetric against the positions of atoms. This equation of motion is an interpolation between the master equation corresponding to the most co-operative decay ($\beta = 0$) and the one associated to an incoherent decay ($\beta = 1$). The presence of this latter term implies that the expectation value $\langle R^2 \rangle$ does not stay constant during the decay process, except for $\beta = 0$. The equations of motion for the matrix element of the relevant part of $\varrho(t)$ are given and the transition probabilities between different collective spin states agree with those given by DICKE⁽⁹⁾.

Approximate solutions for large N and different β are given in Sect. 3 and compared to previous works. The behaviour of the system in the S.M.E. approach is shown to be very sensitive to the mixing of atomic states with different co-operation numbers. Particularly, this effect leads to delays of the coherent emission of radiation which were not expected in previous works. In Sect. 4 we consider the case of inhomogeneously broadened systems. Finally in Sect. 5 aspects of statistics of the radiation field are considered for suitable detectors set far from the source.

2. — Symmetrized master equation.

2.1. *Model.* — We assume that all atoms have the same transition frequency, ω_0 . Then, within the Born-Markoff approximations, the equation of motion

⁽⁹⁾ R. H. DICKE: in *Quantum Electronics, III*, edited by P. GRIVET and N. BLOEMBERGER (New York, N.Y., 1964), p. 35.

for the reduced atomic density operator $\varrho(t)$ is ^(3b,4a)

$$(2.1) \quad \frac{d\varrho}{dt} = -\frac{1}{2} \sum_{i,j} \Gamma_{ij} \{R_i^+ R_j^- \varrho - R_j^- \varrho R_i^+\} + \text{h.c.} = A\varrho,$$

where

$$(2.2) \quad \Gamma_{ij} = \frac{2\pi^2 \omega_0^3}{\hbar(2\pi)^3} \int d^3k \frac{\delta(\omega - \omega_0)}{\omega} \left\{ \mu^2 - \left(\frac{\boldsymbol{\mu} \cdot \mathbf{k}}{|\mathbf{k}|^2} \right)^2 \right\} \exp[i\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)].$$

When the linear dimensions of the sample are larger than the radiation wavelength, the Γ_{ij} are generally different for any pair (i, j) , so that the atoms become distinguishable through their interaction with the electromagnetic field. Then as previously seen, the square length of the angular momentum is not a constant of the motion.

We follow DICKE ⁽¹⁾ and use the standard basis $\{|\eta rm\rangle\}$ of the collective spin operators: m stands for the eigenvalue of the collective energy operator $R_3 = \sum_j R_{3j}$, r is the co-operation number defined by

$$(2.3) \quad R^2 |\eta rm\rangle = r(r+1) |\eta rm\rangle,$$

and η labels a state of given (r, m) which has a degeneracy of order

$$(2.4) \quad n_r = \frac{N!(2r+1)}{(N/2-r)!(N/2+r+1)!}.$$

The set $|\eta rm\rangle$ ($\eta = 1, \dots, n_r$) for given (r, m) forms an irreducible representation of the group of the permutations ⁽¹⁰⁾ over the N spin variables; for any permutation S we have

$$(2.5) \quad S|\eta rm\rangle = \sum_{\eta'=1}^{n_r} S_{\eta, \eta'}^r |\eta' r m\rangle.$$

Let us recall some properties of finite groups which will be useful in the derivation of the model.

Any operator A which acts upon the spin variables is symmetric if it obeys the relation

$$(2.6a) \quad A = \frac{1}{N!} \sum_s S^{-1} A S.$$

By using the orthogonality relations of finite groups ⁽¹⁰⁾ it is easy to show

⁽¹⁰⁾ M. HAMERMERSH: in *Group Theory and Its Application to Physical Problems* (Reading, Mass., 1962).

from eq. (2.6a) that A can be expressed as

$$(2.6b) \quad A = \sum_{r,m,m',\lambda} \frac{1}{n_r} \langle \lambda r m | A | \lambda r m' \rangle \sum_{\eta} |\eta r m \rangle \langle \eta r m' |.$$

Then the matrix elements of any symmetric spin operator are diagonal on r and η ; furthermore they are independent of η . Now let us consider a spin operator B which does not satisfy the relation (2.6). We can draw out the symmetric part of B with the help of the symmetrization operator $(^{10})$ P , defined by

$$(2.7a) \quad PB = \frac{1}{N!} \sum_s S^{-1} B S.$$

It is obvious from orthogonality relations that PB can be also expressed as

$$(2.7b) \quad PB = \sum_{\substack{r,m,m',\lambda}} \frac{1}{n_r} \langle \lambda r m | B | \lambda r m' \rangle \sum_{\eta} |\eta r m \rangle \langle \eta r m' |,$$

and that

$$\text{Tr } PB = \text{Tr } B.$$

When the standard basis $\{|\eta r m \rangle\}$ is used to study the master equation (2.1) the matrix of the density operator $\varrho(t)$ is not diagonal on r and η . Then, $\varrho(t)$ is not symmetric with respect to the permutations of the spin variables. We always can split $\varrho(t)$ in the following way:

$$(2.8) \quad \varrho(t) = P\varrho(t) + (1 - P)\varrho(t),$$

where P is the symmetrization operator defined by eq. (2.7a). By definition, the operator $P\varrho$ is symmetric and satisfies

$$(2.9) \quad \left\{ \begin{array}{l} P\varrho = \sum_{\substack{r,m,m',\lambda}} \frac{1}{n_r} \langle \lambda r m | \varrho | \lambda r m' \rangle \sum_{\eta} |\eta r m \rangle \langle \eta r m' |, \\ \text{Tr } P\varrho = \text{Tr } \varrho = 1. \end{array} \right.$$

Later on, the operator $P\varrho$ will be denoted ϱ_s and the nonsymmetric part $(1 - P)\varrho$, which verifies

$$P(1 - P)\varrho = 0,$$

will be ϱ_{NS} . When we apply the operators P and $1 - P$ to the master equation (2.1), we obtain coupled equations of motion for ϱ_s and ϱ_{NS} . Let A_s , the

symmetrized part of A , be defined by PA ; we have

$$(2.10) \quad \frac{d\rho_s}{dt} = A_s \rho_s + P(A \rho_{NS}),$$

$$(2.11) \quad \frac{d\rho_{NS}}{dt} = (A - A_s) \rho_s + A \rho_{NS} - P(A \rho_{NS}).$$

The second equation can be also written

$$(2.12) \quad \frac{d\rho_{NS}}{dt} = (A - A_s) \rho_s + A_{NS} \rho_{NS}.$$

Introducing the solution of $\rho_{NS}(t)$ into eq. (2.10), we obtain

$$(2.13) \quad \begin{aligned} \frac{d}{dt} \rho_s(t) = & A_s \rho_s(t) + P(A \exp[A_{NS}t] \rho_{NS}(0)) + \\ & + P\left(\int_0^t du A \exp[A_{NS}u] (A - A_s) \rho_s(t-u)\right) \end{aligned}$$

with

$$(2.14) \quad \rho_{NS}(t) = \exp[A_{NS}t] \rho_{NS}(0) + \int_0^t du \exp[A_{NS}u] (A - A_s) \rho_s(t-u).$$

The contribution from $\rho_{NS}(t)$ in the equation of motion of $\rho_s(t)$ contains two terms. The first one depends on the initial atomic state; the second term is at least of fourth order in the interaction V , and will be later neglected against $A_s \rho_s$, within an accuracy of order $1/N$ (see Appendix B). Now, if we assume that all atoms have been equivalently excited, *i.e.* $\rho_{NS}(0) = 0$, we obtain for $\rho_s(t)$ the following master equation:

$$(2.15) \quad \frac{d\rho_s}{dt} = A_s \rho_s.$$

Then, $\rho_s(t)$, which is defined by the relations (2.10), will be the relevant part of $\rho(t)$. The symmetrized master equation (S.M.E.) given by eq. (2.15) will describe the behaviour of an atomic system which has been prepared in a symmetric state. This condition, which is essential for the validity of the S.M.E., will be discussed in Subsect. 2'2.

Later, the time will be in units of Γ^{-1} , and Γ_{ij} will stand for Γ_{ij}/Γ . Now let us calculate A_s . It is easy to show that, from eqs. (2.7a) and (2.7b) (Appendix C),

$$(2.16) \quad P\left(\sum_{i,j} \Gamma_{ij} R_i^+ R_j^-\right) = (1 - \beta) R^+ R^- + \beta \sum_i R_i^+ R_i^-,$$

where the parameter β is defined by

$$(2.17) \quad \beta = \frac{N}{N-1} \left[1 - \frac{1}{N^2} \sum_{i,j} \Gamma_{ij} \right].$$

Therefore the Liouvillian operator A_s will be defined by

$$(2.18) \quad A_s \varrho_s = (1 - \beta) A_c \varrho_s + \beta A_I \varrho_s,$$

where A_c is the Liouvillian operator for the most co-operative case of emission

$$(2.19) \quad A_c \varrho_s = -\frac{1}{2} \{ R^+ R^- \varrho_s - R^- \varrho_s R^+ + \text{h.c.} \},$$

and A_I the incoherent Liouvillian operator for independently decaying atoms (^{2c})

$$(2.20) \quad A_I \varrho_s = -\frac{1}{2} \sum_i \{ R_i^+ R_i^- \varrho_s - R_i^- \varrho_s R_i^+ + \text{h.c.} \}.$$

So, the master equation for $\varrho_s(t)$ represents an interpolation between the two limiting cases of spontaneous emission corresponding respectively to $\beta = 0$ and $\beta = 1$. Due to A_I , it follows that $\langle R^2 \rangle$ will not be a constant of the motion. Hence the fundamental property of the problem, *i.e.* $[R^2, H^-] \neq 0$, is not altered in the S.M.E. approach.

From eqs. (2.19) and (2.20) we can express the equation of motion for matrix elements of ϱ_s . By using the Wigner-Eckart theorem we obtain

$$(2.21) \quad \frac{d\varrho_{r,m}}{dt} = -A_{r,m} \varrho_{r,m} + B_{r,m+1} \varrho_{r,m+1} + C_{r-1,m+1} \varrho_{r-1,m+1} + D_{r+1,m+1} \varrho_{r+1,m+1},$$

where $\varrho_{r,m}$ stands for $\langle rm | \varrho_s | rm \rangle$ and

$$(2.22) \quad \left\{ \begin{array}{l} A_{r,m} = v_{r,m}(1 - \beta) + \beta(N/2 + m), \\ B_{r,m} = v_{r,m} \left[(1 - \beta) + \beta \frac{N/2 + 1}{2r(r+1)} \right], \\ C_{r,m} = \beta \frac{N/2 + r + 2}{2(r+1)(2r+3)} (r - m + 1)(r - m + 2), \\ D_{r,m} = \beta \frac{N/2 - r + 1}{2r(2r-1)} (r + m)(r + m - 1), \\ v_{r,m} = (r + m)(r - m + 1). \end{array} \right.$$

A state $|rm\rangle$ is populated by the states

$$|r+1, m+1\rangle, \quad |r, m+1\rangle, \quad |r-1, m+1\rangle$$

with respective transition rates

$$n_{r+1} D_{r+1, m+1}, \quad n_r B_{r, m+1} \quad \text{and} \quad n_{r-1} C_{r-1, m+1}.$$

When taking $N=6$, we find again the transition rates given by DICKE⁽⁹⁾.

The respective weights of co-operative and incoherent processes will depend on the quantities

$$(2.23) \quad \begin{cases} a_c = n_{rm}(1 - \beta), \\ a_i = \beta \left(\frac{N}{2} + m \right). \end{cases}$$

The parameter $1 - \beta$ defined by eq. (2.17) is generally a small one. The quantity

$$(2.24) \quad \alpha = \frac{1}{N^2} \sum_{i,j} \Gamma_{ij} \sim 1 - \beta,$$

which can be calculated for samples with large concentrations, is quite small. For large Fresnel number (a condition which is always satisfied in the optical-frequency range), it has been obtained^(3e,11) that

$$(2.25) \quad \alpha \sim \frac{\lambda^2}{A},$$

where λ is the radiation wavelength and A the sample section. Then, with $A = 1 \text{ cm}^2$ and $\lambda \approx 10^{-4}, 10^{-5} \text{ cm}$, we have

$$\alpha = 1 - \beta \sim 10^{-8}, 10^{-10}.$$

Therefore, for the decay of highly excited states, *i.e.* for large values of r and m , the incoherent decay will prevail against the co-operative decay. When the system approaches the zero-energy state, we have to compare

$$a_c = r(r+1)(1 - \beta)$$

with

$$(2.26) \quad a_i = \frac{N}{2} \beta.$$

For large r , the co-operative decay would be important only if $N(1 - \beta)$ is much larger than unity ($N(1 - \beta)$ is equivalent to the factor s of ERNST and STEHLE^(3a)

⁽¹¹⁾ I. D. ABELLA, N. A. KURNIT and S. R. HARTMAN: *Phys. Rev.*, **141**, 391 (1966).

and to Rehler and Eberly's ^(3d) for $\mathbf{k}_0 = 0$). But for small r , the incoherent decay always prevails. Therefore, in the region $m = 0$, a fast decay of the atomic system will appear only if $N(1 - \beta)$ is much greater than unity and if the states $|r, 0\rangle$ with large r are strongly populated, or, equivalently, when the expectation value $\langle R^2 \rangle$ is large.

We close this Subsection by giving the equations of motion for the expectation values of atomic operators in the S.M.E. approach.

First let us consider an operator A invariant under any permutation S . As the operation trace is a symmetrization operation, one has

$$(2.27) \quad \text{Tr } A \varrho = \text{Tr } P(A \varrho) = \text{Tr } A \varrho_S.$$

So, when A is symmetric, only the knowledge of ϱ_S is necessary to calculate its expectation value. Now, consider a nonsymmetric operator B . Then

$$(2.28) \quad \text{Tr } B \varrho = \text{Tr } (PB) \varrho_S + \text{Tr } P(B \varrho_{\text{NS}}).$$

As $\varrho_{\text{NS}}(0)$ is taken equal to zero, $\varrho_{\text{NS}}(t)$ is at least of order V^2 against $\varrho_S(t)$ (eq. (2.14)). So, the most important contribution arises from ϱ_S , and eq. (2.28) can be approximated by

$$(2.29) \quad \text{Tr } B \varrho \sim \text{Tr } (PB) \varrho_S.$$

Then by using the master equation (2.18), the equation of motion for the expectation value of *any* operator will be

$$(2.30) \quad \frac{d}{dt} \langle A \rangle = -\frac{1}{2} \{ (1 - \beta) \text{Tr} [A, R^+] R^- \varrho_S + \beta \text{Tr} \sum_i [A, R_i^+] R_i^- \varrho_S + \text{h.c.} \}.$$

For $\langle R^2 \rangle$ we have

$$(2.31) \quad \frac{d}{dt} \langle R^2 \rangle = -\beta [2 \langle R^2 \rangle + N(\langle R_3 \rangle - 1) - \langle R^+ R^- \rangle].$$

The expectation values $I_l = \langle (R^+)^l (R^-)^l \rangle$ and $W_{k,l} = \langle (R^+)^l R_3^k (R^-)^l \rangle$ obey a hierarchy of equations, bounded in l owing to initial conditions, but infinite in k ,

$$(2.32) \quad \left\{ \begin{array}{l} \frac{d}{dt} I_l = 2(1 - \beta) \left[\frac{l(l-1)}{2} I_l + l W_{1,l} \right] - \beta l I_l, \\ \frac{d}{dt} W_{1,l} = 2(1 - \beta) \left[\frac{l(l-1)}{2} W_{1,l} + l W_{2,l} - \frac{1}{2} I_{l+1} \right] - \\ \qquad \qquad \qquad - \beta \left[\frac{N}{2} I_l + (l+1) W_{1,l} \right], \\ \frac{d}{dt} W_{2,l} = \dots \end{array} \right.$$

In the general case the hierarchy of equations cannot be exactly solved. An approach to know the behaviour of the system can be attempted by truncating the set (2.32) beyond a suitable order, and by neglecting the quantum correlations beyond this order. For small samples ($\beta = 0$) the hierarchy (2.32) can be reduced to a closed set of equations⁽¹²⁾ for I_i and $W_{1,i}$ by using the invariance of $\langle R^2 \rangle$.

2'2. Discussion of S.M.E. — The symmetrized master equation (2.18) is valid within an accuracy $1/N$, when the atomic system has been prepared in a symmetric state, *i.e.* when its initial density operator is given by

$$(2.33) \quad \varrho(0) = \sum_{\substack{r,m \\ m'}} \langle rm | \varrho(0) | rm' \rangle \sum_{\eta} |\eta rm\rangle \langle \eta rm'|.$$

We will distinguish two ways for the excitation of the atoms.

First consider the case of an incoherent excitation^(1,2c) for which each atom interacts with a particle of the field (photon, electron, ...). For a field constructed with identical particles, one can assume

$$(2.34) \quad \begin{cases} \varrho_{++}^i = \varrho_{++}, \\ \varrho_{--}^i = \varrho_{--}, \\ \varrho_{+-}^i = 0. \end{cases}$$

And with

$$(2.35) \quad |\eta rm\rangle = \sum_{\{n_i\}\{n_j\}} C(\eta, r, m; \{n_i\} \{n_j\}) \delta_{2m, \{n_i^+\} - \{n_j^-\}} |\{n_i^+\} \{n_j^-\}\rangle,$$

where the initial energy of system is

$$m = \frac{1}{2} \sum \varrho_{++}^i - \varrho_{--}^i,$$

we obtain from (2.34)

$$(2.36) \quad \langle \eta rm | \varrho(0) | \eta rm \rangle = \prod_{i=1}^{N/2+m} \varrho_{++}^i \prod_{j=1}^{N/2-m} \varrho_{--}^j = \langle m | \varrho | m \rangle.$$

Then the initial density operator is symmetric:

$$(2.37) \quad \varrho(0) = \langle m | \varrho | m \rangle \sum_r |\eta rm\rangle \langle \eta rm|.$$

Now let us consider that the atoms have been excited by an intense pulse

⁽¹²⁾ G. OLIVER and A. TALLET: *Phys. Rev. A*, **7**, 1061 (1973).

with wave vector $\mathbf{k}_0$ and area θ . By neglecting all relaxation processes during the excitation, and assuming the inverse of the nutation frequency much smaller than L/c , such a pulse leaves each atom in a superposition of its upper and lower states. After the passage of the pulse, the wave function φ_k of the k -th atom is ^(3d)

$$(2.38) \quad |\varphi_k\rangle = \sin \frac{\theta}{2} |+\rangle_k + \exp[-i\mathbf{k}_0 \cdot \mathbf{r}_k] \cos \frac{\theta}{2} |-\rangle_k.$$

Strictly apart from the value $\theta = \pi$, the initial density operator will not be symmetric. But it will be diagonal in r and η , in the standard basis $\{|\eta r m; \mathbf{k}_0\rangle\}$ of $R_{k_0}^2$ and R_3 . Nevertheless, by using this basis, a method similar to the symmetrization described above would lead one to consider that any operator

$$\exp[i\mathbf{k}_0 \cdot (\mathbf{r}_i - \mathbf{r}_j)] R_i^+ R_j^-$$

is independent of positions $\mathbf{r}_i$ and $\mathbf{r}_j$. Such a model is not valid since it would privilege *a priori* the direction $\mathbf{k}_0$.

For large N , one can assume ^(2c) that a coherent excitation leaves the system in an eigenstate of R_3 . So, for $\theta \sim \pi$, *i.e.* $\langle R_3(0) \rangle = m \sim N/2$, only the states $|\eta r m\rangle$, with $r \geq m$, are populated. The irrelevant part $\varrho_{\text{NS}}(0)$ will be negligible against $\varrho_s(0)$, so that the S.M.E. approach can be still used. But, for a coherent excitation which leaves the system in the superradiant state $|(N/2) m = 0; \mathbf{k}_0\rangle$, the S.M.E. is no longer valid. Such an excitation condition will not be considered later.

REHLER and EBERLY ^(3d) assume that, after the passage of an intense pulse with a wave vector $\mathbf{k}_0$, each atom will decay, by retaining the same configuration as eq. (2.32). The decay process is then described by a single parameter $\theta(t)$ which obeys the law of energy conservation. As each atom decays equivalently with the same $\theta(t)$, it involves that the atomic system will stay in the state $|(N/2) m; \mathbf{k}_0\rangle$. Consequently with regard to the standard basis $\{|\eta r m; \mathbf{k}_0\rangle\}$, the co-operation number of the system will be constant. However for the spontaneous-emission case the expectation value $R_{k_0}^2$ is not *a priori* a constant of the motion as we can see from eq. (1.1) or eq. (2.1). Their description leads also to a decay process with a characteristic time depending on the excitation wave vector $\mathbf{k}_0$. This seems to be *a priori* a rough approximation: indeed any eigenvalue of an operator must be independent of the chosen complete basis. The model of REHLER and EBERLY seems to be more appropriate to describe the co-operative decay of atoms contained in a cavity and interacting with a single mode. Maybe, however, their model would be valid when the atoms have been excited into the superradiant state; for short times the decay is so fast that the atoms may retain the configuration of the excitation process (eq. (2.32)).

Let us point out that in the master equation (2.1) frequency shift terms have

been neglected. This approximation is consistent with the hypothesis (2.33). Indeed, from the S.M.E. (eq. (2.15)), it is easy to see that the behaviour of the system is completely determined by the diagonal elements of ϱ_s . Therefore there are no contributions of frequency shift terms.

3. - Approximate solutions of S.M.E.

To characterize the spontaneous emission we are particularly interested in the behaviour of the energy of the system $\langle R_3 \rangle$, the radiation rate defined as $-(d/dt)\langle R_3 \rangle$ and also with the expectation value $\langle R^2 \rangle$. From eq. (2.31) and from the hierarchy (2.32), we can see that the expectation values $\langle R^2 \rangle$ and $\langle R_3 \rangle$ depend on the expectation value $\langle R^+ R^- \rangle$. So, to obtain the solution for $\langle R_3 \rangle$ we have to solve the coupled equations

$$(3.1) \quad \begin{cases} \frac{d}{dt} \langle R_3 \rangle = -(1 - \beta) \langle R^+ R^- \rangle - \beta \left\langle \frac{N}{2} + R_3 \right\rangle, \\ \frac{d}{dt} \langle R^+ R^- \rangle = 2(1 - \beta) \langle R^+ R_3 R^- \rangle - \beta \langle R^+ R^- \rangle. \end{cases}$$

And knowledge of $\langle R_3 \rangle$ and $\langle R^+ R^- \rangle$ leads to the solutions for $\langle R^2 \rangle$.

To solve the coupled equations (3.1), the expectation value $\langle R^+ R_3 R^- \rangle$ will be approximated by the product $\langle R^+ R^- \rangle \langle R_3 \rangle$, by neglecting the quantum correlations beyond the second order. First consider the case $\beta = 0$. Formally it is not valid to neglect the quantum correlations at any time. Indeed it has been shown ⁽¹²⁾ that, for *any* initial atomic state, quantum effects appear for long times, in the tail of the radiated pulse, *i.e.* when the system approaches the ground state. The correlation functions $\langle R^{+k} R_3^l R^{-k} \rangle / \langle R^+ R^- \rangle^k \langle R_3 \rangle^l$ are increasing beyond unity for these times, but any expectation value $\langle R^{+k} R_3^l R^{-k} \rangle$ is vanishing. Large fluctuations express only the fact that $\langle R^{+k} R_3^l R^{-k} \rangle$ vanishes slower than the product $\langle R^+ R^- \rangle^k \langle R_3 \rangle^l$. Then, the decorrelation beyond the second order into eq. (3.1) should not essentially affect the behaviour of the expectation value $\langle R^+ R^- \rangle$ and $\langle R_3 \rangle$. For the opposite case, $\beta = 1$, we obtain easily from eq. (2.32)

$$\langle R^+ R_3 R^- \rangle = \langle R^+ R^- \rangle \langle R_3 \rangle - \langle R^+ R^- \rangle \exp[-\Gamma t],$$

when we assume that at $t=0$ the system is in a pure state. So, for $\beta=1$, one can neglect the third-order correlations within an accuracy of order $1/N$.

Then, as long as we are concerned only with the behaviour of $\langle R_3 \rangle$ and $\langle R^2 \rangle$, the approximation

$$\langle R^+ R_3 R^- \rangle \sim \langle R^+ R^- \rangle \langle R_3 \rangle$$

seems to be reasonable.

With the definitions

$$(3.2) \quad \begin{cases} x = \frac{1}{N} \left[\langle R_s \rangle + \frac{N}{2} \right], \\ i = \frac{\langle R^+ R^- \rangle}{N}, \\ b = N(1 - \beta), \end{cases}$$

eqs. (3.1) become

$$(3.3) \quad \begin{cases} \frac{dx}{dt} = - \left[\left(1 - \frac{b}{N} \right) x + \frac{b}{N} i \right], \\ \frac{di}{dt} = i \left[2bx - \left(1 + b - \frac{b}{N} \right) \right]. \end{cases}$$

From Subsect. 2'1 we have seen that $1 - \beta = b/N$ is yet very small for samples with large concentrations. Then it will be neglected against unity. So, we have to solve

$$(3.4) \quad \begin{cases} \frac{dx}{dt} = - \left[x + \frac{b}{N} i \right], \\ \frac{di}{dt} = i[2bx - (1 + b)]. \end{cases}$$

As long as $(b/N)i(t)$ is very small compared to unity, it can be neglected in eq. (3.4). Then, the solutions are

$$(3.5) \quad \begin{cases} x = x(0) \exp[-t], \\ i = i(0) \exp[-(b+1)t] \exp[2bx(0)[1 - \exp[-t]]]. \end{cases}$$

The condition

$$(3.6) \quad \frac{b}{N} i(t) \ll 1$$

gives an estimate of the validity of the solutions (3.5), as a function of b . For these solutions, $i(t)$ reaches a maximum for such a time t_M

$$(3.7) \quad \left\{ \begin{array}{l} x(0) \exp[-t_M] = \frac{1}{2} \frac{b+1}{b}, \\ \exp[-t_M] = \frac{1}{2x(0)}, \end{array} \right. \text{ or }$$

since we search the upper limit of b for which eq. (3.6) is met. Then

$$(3.8) \quad i_M = i(0) \left[\frac{1}{2x(0)} \right]^{b+1} \exp \left[2b \left[x(0) - \frac{1}{2} \right] \right],$$

and the inequality (3.6) leads to

$$(3.9) \quad [2x(0) - 1 - \log 2x(0)]b \ll \log \frac{2Nx(0)}{bi(0)}.$$

If we assume $x(0) = i(0) \approx 1$, eq. (3.9) becomes

$$(3.10) \quad b + 3 \log b \ll 3 \log 2N.$$

Then, when b satisfies inequality (3.10), the solution (3.5) will be valid for any time. For example, with $N = 10^{10}$, the inequality (3.10) gives $b \leq 50$ and $N = 10^{17}$ leads to $b \leq 100$.

A first conclusion can be drawn from these results. We see that the decay process is incoherent with respect to the energy and the radiation rate for a large range of b , depending on N . This result disagrees with the previous works of REHLER and EBERLY^(3d) and of ERNST and STEHLE^(3e). In their models, the behaviour of the system deviates from the incoherent one as soon as $b \neq 0$, and yet for $b = 2$ the normalized radiation rate reaches a maximum greater than unity. It is clear that in the previous works the co-operative effects

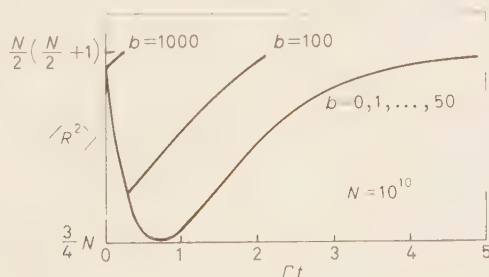


Fig. 1. — The square length of the collective angular momentum $\langle R^2 \rangle$ as a function of Γt with $N = 10^{10}$ and different b . For $b = 0, 1, \dots, 50$, the behaviour of $\langle R^2 \rangle$ is typical of independently decaying atoms. For larger b , the minimum of $\langle R^2 \rangle$ increases as the co-operative effects become important.

are favoured because the co-operation number is taken constant. In the S.M.E. approach, the time evolution of $\langle R^2 \rangle$ can be seen in Fig. 1, where eqs. (3.3) and (2.31) have been numerically integrated with $N = 10^{10}$. For values of b which obey the condition (3.10), i.e. for an incoherent process, $\langle R^2 \rangle$ reaches

near the minimum value $3N/4$ corresponding to

$$\langle R^2 \rangle = \sum_i \langle \mathbf{R}_i \cdot \mathbf{R}_i \rangle.$$

This value is reached when the energy of the system is zero, *i.e.* when the atoms are equally distributed among the states $|r, 0\rangle$.

The behaviour of normalized energy is shown in Fig. 2 and 3, where eqs. (3.3) have been numerically computed for different values of b and N . For short times such that $(b/N)i(t)$ is small against 1, the behaviour is always given by eq. (3.5). But when $i(t)$ is growing, $x(t)$ rapidly decreases and finally for

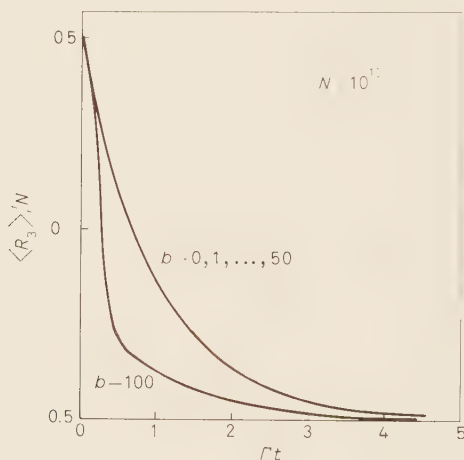


Fig. 2. — Energy of the system, normalized to N as a function of Γt , with $N = 10^{10}$. For $b = 0, 1, \dots, 50$, the curve is the decaying exponential $\exp[-\Gamma t]$. For $b = 100$, fast decay appears.

long times decreases again as $\exp[-t]$. This behaviour is shown in Fig. 2 with $N = 10^{10}$ and $b = 100$ as a function of the time Γt . It can be compared to the incoherent decay numerically obtained for $b < 50$, concurring with analytical approximate solutions (3.5). In Fig. 3, where the horizontal scale is in units of $(b\Gamma)^{-1}$, we can compare the variation of normalized energy for two values of N with the same b ($b = 10^3$). From the above discussion, it is clear that co-operative effects appear at times depending on N , due to the importance of $(b/N)i(t)$. This result also disagrees with previous works (^{3a,3d}) in which $x(t)$ is only a function of the parameter b . During the fast decay, the curves are parallel for $N = 10^{10}$ and $N = 10^{14}$, and also parallel to the semi-classical curve (^{3d}) (dashed line). Also the time t , at which the fast decay starts, can be determined from eqs. (3.5) and (3.6). For large b , $\langle R^2 \rangle$ is nearly constant.

In conclusion, the S.M.E. leads to a description for the spontaneous emission

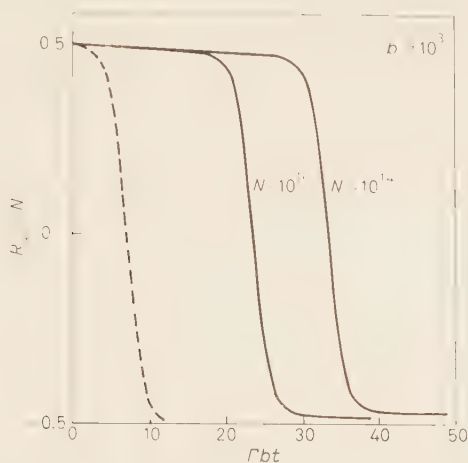


Fig. 3. — Energy of the system, normalized to N as a function of Γbt for the same b , but with different N . The dashed line represents the energy normalized to N , calculated in the semi-classical model (^{3d}).

of highly excited atoms which is quite different from that previous models, particularly in the range of low concentrations. With the large concentrations generally encountered in experiments on ruby (¹¹) or SF_6 (¹³), the parameter b is very great. For these cases the contribution of the incoherent process gives only a retardation for the co-operative emission of radiation.

4. — Inhomogeneous-broadening case.

When the atomic system is inhomogeneously broadened, the Born-Markoff master equation is also given by eq. (2.1) but with

$$(4.1) \quad \Gamma_{ij} = \frac{2\pi^2 \omega_0^2}{\hbar(2\pi)^3} \int d^3 k \frac{\delta(\omega - \omega_j)}{\omega} \left\{ \mu^2 - \frac{(\boldsymbol{\mu} \cdot \mathbf{k})^2}{|\mathbf{k}|^2} \right\} \exp[i\mathbf{k}(\mathbf{r}_i - \mathbf{r}_j) - i(\omega_i - \omega_j)t].$$

Assuming that the frequency distribution $g(\Delta\omega)$ of the full width $(T_2^*)^{-1}$ is such that

$$(4.2) \quad \frac{L}{c} \ll T_2^*,$$

we can neglect in eq. (4.1) the dependence of $\mathbf{r}_i - \mathbf{r}_j$ on $\omega_j - \omega_0$. So when decoupling the dependence on positions and on inhomogeneous broadening in Γ_{ij} is possible, the parameter b defined by eq. (2.17) and eq. (3.2) will

(¹³) C. K. N. PATEL and R. E. SLUSHER: *Phys. Rev. Lett.*, **20**, 1087 (1968).

be multiplied by the integral

$$\left| \int_{-\infty}^{+\infty} g(\Delta\omega) \exp [i \Delta\omega t] d(\Delta\omega) \right|^2.$$

For a Lorentzian shape of $g(\Delta\omega)$, normalized to unity, then the factor $b = N(1 - \beta)$ will be replaced in all above equations by

$$(4.3) \quad \tilde{b}(t) = b \exp \left[-\frac{t}{T_2^*} \right].$$

Calculations performed in Sect. 3 can be made again by including $\tilde{b}$ in place of b . It is clear that co-operative effects will not be favoured by the decaying exponential factor $\exp [-t/T_2^*]$. So, let us consider the effect of inhomogeneous broadening on the coupled equations (3.4). One has the solutions

$$(4.4) \quad \left\{ \begin{array}{l} x = x(0) \exp [-\Gamma t], \\ i(t) = i(0) \exp \left[\Gamma b \left[\frac{2x(0)}{\Omega} - T_2^* \right] - \right. \\ \qquad \qquad \qquad \left. - \Gamma t - b \Gamma \left[\frac{2x(0)}{\Omega} \exp [-\Omega t] - T_2^* \exp \left[-\frac{t}{T_2^*} \right] \right] \right], \\ \Omega = \Gamma + \frac{1}{T_2^*}, \end{array} \right.$$

as long as

$$(4.5) \quad \frac{\tilde{b}(t)}{N} i(t) \ll 1.$$

Henceforth, we will assume that the radiative lifetime Γ^{-1} is much smaller than T_2^* . This condition is met for forbidden transitions corresponding to materials used in most experiments of nonlinear optics (^{11,13}). Then, with $\Omega^{-1} = T_2^*$, we have

$$(4.6) \quad i(t) \sim i(0) \exp \left[\Gamma b T_2^* [2x(0) - 1] \left[1 - \exp \left[-\frac{t}{T_2^*} \right] \right] \right].$$

The time t_M , for which $i(t)$ reaches a maximum, is approximatively given by

$$\exp \left[-\frac{t_M}{T_2^*} \right] = \frac{1}{b[2x(0) - 1]}$$

with

$$(4.7) \quad i_M \approx i(0) \exp [\Gamma b T_2^* [2x(0) - 1]].$$

So, by taking $x(0) = i(0) \approx 1$, the condition (4.5) becomes

$$(4.8) \quad b \ll \frac{1}{\Gamma T_2^*} \log N.$$

And as long as b verifies inequality (4.8), the solutions for $x(t)$ and $i(t)$ are given by eq. (4.4). If we compare eq. (4.8) to eq. (3.10), we see that incoherent decay of inhomogeneously broadened atoms is expected even for large values of b , *i.e.* for large concentrations of atoms. As an example, let us consider the characteristic data in experiments on ruby ⁽¹¹⁾ (we choose the case of solid samples, because the condition (4.2) is generally met)

$$T_2^* \approx 10^{-10} \text{ s}, \quad \Gamma \approx 10^3 \text{ s}^{-1}, \quad \lambda \approx 7 \cdot 10^{-5} \text{ cm}.$$

So, for a unit volume (1 cm^3), with $b/N \approx 10^{-10}$, the inequality (4.8) is satisfied for $N \leq 10^{18}$. Then, even for large concentrations, the inhomogeneous broadening can quench the co-operative emission of radiation ⁽¹⁴⁾.

AGARWAL ^(4b) studied the effect of inhomogeneous broadening on the co-operative emission of small samples (*i.e.* $b = N$). As he considered that $\langle R^2 \rangle$ was a constant of the motion, with

$$\langle R^2 \rangle = \frac{N}{2} \left(\frac{N}{2} + 1 \right)$$

his result ^(4b) disagrees with our conclusions. In effect, in our model, taking $b = N$, we should obtain

$$x(t) = \exp[-\Gamma t],$$

valid for

$$(4.9) \quad \Gamma T_2^* \ll \frac{\log N}{N}.$$

As in the case of small volumes ($V \sim \lambda^3$) N is quite small, the condition (4.8) is always met, and the behaviour of the system would be typical of an incoherent process.

5. — Detection of the radiation field.

It is well known that with the help of the Heisenberg equations, the electromagnetic-field operators can be expressed in terms of atomic operators. The

⁽¹⁴⁾ $\Gamma b T_2^*$ can be identified with τ_2^*/τ_s , where τ_s is the «superradiant» lifetime defined by GONCALVES *et al.* (*Phys. Rev.*, **188**, 576 (1969)). $\Gamma b T_2^*$ is also proportional to αL , where α^{-1} is the linear absorption coefficient.

positive-frequency part of the potential vector operator, at position $\mathbf{R}$ and time t , is written as

$$(5.1) \quad A^+(\mathbf{R}, t) = A_0^+(\mathbf{R}, t) - \epsilon \sum_j \frac{\exp[ik_0|\mathbf{R} - \mathbf{r}_j|]}{|\mathbf{R} - \mathbf{r}_j|} R_j^-(t - |\mathbf{R} - \mathbf{r}_j|/c).$$

$A_0^+(\mathbf{R}, t)$ is the free potential vector operator, and $\omega_0 = k_0 c$ is the transition frequency of any atom. (Here we neglect the inhomogeneous broadening.) So, when the field is in the vacuum state at $t = 0$, the field intensity is

$$(5.2) \quad \langle A^-(\mathbf{R}, t) A^+(\mathbf{R}, t) \rangle = |\epsilon|^2 \sum_{j,l} \frac{\exp[-ik_0|\mathbf{R} - \mathbf{r}_l| + ik_0|\mathbf{R} - \mathbf{r}_j|]}{|\mathbf{R} - \mathbf{r}_l||\mathbf{R} - \mathbf{r}_j|} \langle R_l^+(\tau_l) R_j^-(\tau_j) \rangle,$$

where $\tau = t - |\mathbf{R} - \mathbf{r}|/c$ is the retarded time. We will assume later that detectors are set far from the source, so that

$$(5.3) \quad \exp[ik_0|\mathbf{R} - \mathbf{r}_j| - ik_0|\mathbf{R} - \mathbf{r}_l|] \sim \exp\left[ik_0 \frac{\mathbf{R}}{|\mathbf{R}|} \cdot (\mathbf{r}_l - \mathbf{r}_j)\right].$$

Then, eq. (3.2) becomes

$$(5.4) \quad \langle A^-(\mathbf{R}, t) A^+(\mathbf{R}, t) \rangle = \frac{\epsilon^2}{|\mathbf{R}|^2} \sum_{j,l} \exp\left[ik_0 \frac{\mathbf{R}}{|\mathbf{R}|} \cdot (\mathbf{r}_l - \mathbf{r}_j)\right] \langle R_l^+(\tau) R_j^-(\tau) \rangle.$$

By neglecting the contributions of the irrelevant part of $\varrho(t)$, the field intensity, normalized to $\epsilon^2/|\mathbf{R}|^2$, is

$$(5.5) \quad \langle A^-(\mathbf{R}, t) A^+(\mathbf{R}, t) \rangle = \theta(\Psi) \langle R^+ R^- \rangle + [1 - \theta(\Psi)] \sum_i \langle R_i^+ R_i^- \rangle.$$

The parameter $\theta(\Psi)$ which gives the directivity of the emission of radiation is

$$(5.6) \quad \theta(\Psi) = \frac{1}{V^2} \left| \int d^3r \exp\left[ik_0 \frac{\mathbf{R}}{|\mathbf{R}|} \cdot \mathbf{r}\right] \right|^2 - \frac{1}{N}.$$

If we assume that the atoms are contained in a cylinder with length L and radius a , then the integration of eq. (3.6) gives ^(3d)

$$(5.7) \quad \theta(\Psi) = 4 \left[\frac{\sin(k_0(L/2) \cos \Psi)}{k_0(L/2) \cos \Psi} \times \frac{J_1(k_0 a \sin \Psi)}{k_0 a \sin \Psi} \right]^2 - \frac{1}{N},$$

where Ψ is the angle between the cylinder axis and the direction of observation. So, for detection far from the source ($|\mathbf{R}| \gg L$) the detected intensity is only a function of Ψ . In eq. (3.5), the term proportional to $\theta(\Psi)$ corresponds to the co-operative emission of radiation, i.e. $\sum_{i \neq j} \langle R_i^+ R_j^- \rangle$, whereas the incoherent

emission is isotropic. When the dimensions of the sample satisfy the relation for large Fresnel number, the detected intensity is maximum in a direction parallel to the cylinder axis. In the case of a disk ($k_0 L \ll 1$), then $\theta(\Psi)$ will be equal to unity for $\Psi = 0$.

The n -fold correlation functions of the radiation field can be expressed in the same way as the intensity, with the help of expectation values of collective atomic operators. For example, the twofold correlation function for two detectors set in $\mathbf{R}_1$ and $\mathbf{R}_2$, such that

$$|\mathbf{R}_1 - \mathbf{R}_2| = R \ll L, \quad \tau_1 = \tau_2 = \tau,$$

can be written as

$$(5.8) \quad G^{(2)}(\mathbf{R}_1, t; \mathbf{R}_2, t) = \text{Tr} \sum_{i,j,k,l} \gamma_{il}(\mathbf{R}_1) \gamma_{jk}(\mathbf{R}_2) P\{R_i^+ R_j^+ R_k^- R_l^-\} \varrho_s(\tau),$$

$$(5.9) \quad \gamma_{il}(\mathbf{R}) = \exp \left[ik_0 \frac{R}{|\mathbf{R}|} (\mathbf{r}_i - \mathbf{r}_j) \right].$$

The application of P is straightforward. One obtains

$$(5.10) \quad G^{(2)}(\mathbf{R}_1, t; \mathbf{R}_2, t) = \frac{(N-4)!}{N!} \sum_{i \neq j \neq k \neq l} \gamma_{il}(\mathbf{R}_1) \gamma_{jk}(\mathbf{R}_2) \sum_{i \neq j \neq k \neq l} \text{Tr} R_i^+ R_j^+ R_l^- R_k^- \varrho_s + \\ + \frac{(N-3)!}{N!} \left(\sum_{i \neq j \neq k} \gamma_{jk}(\mathbf{R}_2) + \gamma_{il}(\mathbf{R}_1) + \gamma_{ik}(\mathbf{R}_1) \gamma_{ji}(\mathbf{R}_2) + \gamma_{jk}(\mathbf{R}_2) \gamma_{ij}(\mathbf{R}_1) \right) \cdot \\ \cdot \text{Tr} \sum_{i \neq j \neq k} R_i^+ R_j^+ R_l^- R_k^- \varrho_s + \frac{(N-2)!}{N!} \left(\sum_{i \neq j} \gamma_{ij}(\mathbf{R}_1) \gamma_{ji}(\mathbf{R}_2) + 1 \right) \text{Tr} \sum_{i \neq j} R_i^+ R_j^+ R_l^- R_k^- \varrho_s,$$

and so on for any correlation function. For $\mathbf{R}_1 = \mathbf{R}_2$, eq. (3.10) becomes

$$(5.11) \quad G^{(2)}(\mathbf{R}, t; \mathbf{R}, t) = \theta^2(\Psi) \langle R^+ R^+ R^- R^- \rangle + 4\theta(\Psi)[1 - \theta(\Psi)] \cdot \\ \cdot \left[\frac{N}{2} \langle R^+ R^- \rangle + \langle R^+ R_3 R^- \rangle \right] + 2[1 - \theta(\Psi)]^2 \left[\left\langle \left(\frac{N}{2} + R_3 \right)^2 \right\rangle - \left\langle \frac{N}{2} + R_3 \right\rangle^2 \right].$$

This expression is to be compared with

$$(5.12) \quad [G^{(1)}(\mathbf{R}, t)]^2 = \theta^2(\Psi) \langle R^+ R^- \rangle^2 + \\ + 2\theta(\Psi)[1 - \theta(\Psi)] \langle R^+ R^- \rangle \left\langle \frac{N}{2} + R_3 \right\rangle + [1 - \theta(\Psi)]^2 \left\langle \frac{N}{2} + R_3 \right\rangle^2.$$

For small volumes ($\theta(\Psi) \rightarrow 1$) we find again the usual expressions for the correlation functions. In this case the statistic of the radiation field has been

studied particularly with regard to the intensity moments (^{2c,12,15-17})

$$g^l = I_l / (I_1)^l.$$

The moments g^l have initial values depending on the excitation conditions. For all atoms, excited at $t = 0$, the g^l are equal to $l!$ corresponding to Gaussian field statistics. When the atomic system has been prepared in the superradiant state, one has $g^l = 1$. So, if the atoms have been excited into a state with positive energy, the g^l start with values greater than unity. When the atomic system decays, the g^l rapidly decrease and reach unity. These minima are attained when the radiation rate becomes of order N^2 , and the g^l remain equal to unity as long as the radiation is of order N^2 , i.e. in the region of superradiant emission. But, as the system decays further, the g^l begin to increase (^{12,17}). They may reach values exceeding those of Gaussian fields, when the intensity of the radiation field can still be observable. These large values for g^l express the degree of incoherence of the radiation field with regard to the bunching effect, in the right wing of the radiated pulse. Indeed, while the Gaussian field expresses the maximum of disorder with regard to the entropy, there can exist fields more incoherent than Gaussian fields with regard to the bunching effect. We point out that such incoherent fields have been constructed theoretically (¹⁸) and experimentally (¹⁹).

When considering the decay of independent atoms, the first two terms in eq. (5.10) or eq. (5.11) vanish. With the help of eq. (2.32) we find

$$g^2(t) = 2 \frac{\langle (N/2 + R_3)^2 \rangle - \langle N/2 + R_3 \rangle^2}{\langle N/2 + R_3 \rangle^2}.$$

In the limit of large N one obtains

$$g^2(t) = 2$$

whatever the initial conditions. It is easy to show that in this case we find again the Gaussian field properties for *any* correlation function.

In the general case it seems difficult to discuss *a priori* the degree of coherence of the radiation field. It will depend on the relative importance of different terms appearing in eqs. (5.11) and (5.12). Correlation functions will depend on the parameter b which determines the behaviour of the system and also on the position of the detectors.

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APPENDIX A

Discussion on the Born and Markoff approximations.

We consider the system atoms+field with a Hamiltonian given by eq. (1.1). If we assume that at time $t=0$ there are no correlations between the atoms and the field, the reduced density operator of the atomic system is

$$(A.1) \quad \varrho(t) = \sum_{n=0} (-1)^n \int_0^t dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{2n-1}} dt_{2n} \text{Tr}_F [v(t_1) \dots v(t_{2n}) f_0(R) \otimes \varrho(0)]$$

in the interaction representation. $v(t)$ is the Liouvillian operator associated to $V(t)$ where

$$(A.2) \quad V = \sum_{k,j} \alpha_{kj} \exp[-i\mathbf{k} \cdot \mathbf{r}_j] a_k^\dagger R_j^- + \text{h.c.} = \sum_k V_k.$$

Tr_F means we take the trace over the field variables, and $f_0(R)$ is the density operator of the field at time $t=0$.

As pointed out by HAAKE⁽²⁰⁾, two sorts of terms can be identified in the Dyson series (A.1). The first ones, called A_i , are constructed with two elementary processes associated to the same radiation mode at adjacent times, *i.e.* $v_{k_i}(t_i) v_{k_i}(t_{i+1})$. The others, called «overlapping terms», B_i , have at least two elementary processes at no adjacent times. HAAKE has shown that in the Born approximation, *i.e.*

$$(A.3) \quad \frac{d}{dt} \varrho(t) = -\text{Tr}_F \int_0^t dt' v(t) v(t') f_0(R) \otimes \varrho(t'),$$

all overlapping terms B_i are neglected.

Let us consider the fourth-order terms in eq. (A.1). It consists of one term A , namely

$$(A.4) \quad A = \int_0^t dt_1 \dots \int_0^{t_3} dt_4 \sum_{k_1, k_2} \text{Tr}_F \{v_{k_1}(t_1) v_{k_1}(t_2) v_{k_2}(t_3) v_{k_2}(t_4) f_0(R) \otimes \varrho(0)\},$$

and of two overlapping terms

$$(A.5) \quad \left\{ \begin{array}{l} B_1 = \int_0^t dt_1 \dots \int_0^{t_3} dt_4 \sum_{k_1, k_2} \text{Tr}_F \{v_{k_1}(t_1) v_{k_2}(t_2) v_{k_1}(t_3) v_{k_2}(t_4) f_0(R) \otimes \varrho(0)\}, \\ B_2 = \int_0^t dt_1 \dots \int_0^{t_2} dt_4 \sum_{k_1, k_2} \text{Tr}_F \{v_{k_1}(t_1) v_{k_2}(t_2) v_{k_2}(t_3) v_{k_1}(t_4) f_0(R) \otimes \varrho(0)\}. \end{array} \right.$$

⁽²⁰⁾ F. HAAKE: *Zeits. Phys.*, **223**, 353 (1969).

As we are concerned only with the magnitude of these different terms against each other, we will restrict ourselves only to the contributions for two atoms i and j . When the sum over the radiation modes is replaced by an integral, then the Born approximation gives terms with factor of proportionality

$$(A.6) \quad A \propto -\frac{(\Gamma_{ij})^2}{2} \frac{1}{2} (t - 2r_{ij/c})^2 U(t - 2r_{ij/c}),$$

whereas the first overlapping term B_1 gives

$$(A.7) \quad B_1 \propto -\frac{(\Gamma_{ij})^2}{2} \left[\frac{1}{2} (t - r_{ij/c})^2 U\left(t - \frac{r_{ij}}{c}\right) U\left(\frac{2r_{ij}}{c} - t\right) + \right. \\ \left. + \frac{r_{ij}}{c} \left(t - \frac{3}{2} r_{ij/c}\right) U\left(t - \frac{2r_{ij}}{c}\right) \right],$$

the second term B_2 being zero for two atoms. The integration over the radiation modes has been performed by extending the limits of integration over $\omega_1 - \omega_0$ and $\omega_2 - \omega_0$ from $-\infty$ to $+\infty$.

We see that the Born approximation leads to a delay in the co-operative effects between any pair of atoms equal to $2r_{ij/c}$. The exact delay $r_{ij/c}$ is obtained by adding A and B_1 . Then, we conclude that the Born approximation is not an improvement to the Born-Markoff approximations which gives fourth-order terms proportional to

$$-\left(\frac{\Gamma_{ij}}{2}\right)^2 \frac{t^2}{2} U(t).$$

APPENDIX B

Accuracy of S.M.E.

From the exact master equation (2.1), the equation of motion for ϱ_s is given by

$$(B.1) \quad \frac{d\varrho_s}{dt} = P(A\varrho).$$

Then $\varrho_s(t)$ can be expressed as

$$(B.2) \quad \varrho_s(t) = \sum_{n=0}^{\infty} \frac{t^n}{n!} P[A^n \varrho(0)].$$

If we assume that, at time $t=0$, $\varrho(0) = \varrho_s(0)$, eq. (B.2) becomes

$$(B.3) \quad \varrho_s(t) = 1 + \frac{t}{1!} A_s \varrho_s(0) + \frac{t^2}{2!} A_s^2 \varrho_s(0) + \dots + \frac{t^n}{n!} A_s^n \varrho_s(0) + \\ + P(A - A_s)^2 \varrho_s(0) + \dots$$

The first line of eq. (B.3) leads to the S.M.E. given by eq. (2.18). To have an idea of the accuracy of this equation, we can compare in eq. (B.3) the terms proportional to t^2 . As

$$A_s \varrho_s = (1 - \beta)[R^+ R^- \varrho_s - R^- \varrho_s R^+ + \text{h.c.}] + \varrho \left[\sum_i R_i^+ R_i^- \varrho_s - R_i^- \varrho_s R_i^+ + \text{h.c.} \right],$$

one obtains

$$(B.4) \quad (A - A_s) \varrho_s = \sum_{i \neq j} \theta_{ij} [R_i^+ R_j^- \varrho_s - R_j^- \varrho_s R_i^+] + \text{h.c.},$$

where

$$(B.5) \quad \theta_{ij} = \Gamma_{ij} - (1 - \beta).$$

Then we have to compare

$$(B.6) \quad a_s = \left[(1 - \beta)^2 R^+ R^- R^+ R^- + \beta^2 \left(R_3 + \frac{N}{2} \right)^2 + 2\beta(1 - \beta) R^+ R^- \left(\frac{N}{2} + R_3 \right) \right] \varrho_s(0)$$

with

$$(B.7) \quad b_s = P \sum_{i \neq j} \sum_{k \neq l} \theta_{ij} \theta_{kl} R_i^+ R_j^- R_k^+ R_l^- \varrho_s(0),$$

where a_s and b_s have been respectively taken out of $A_s^2 \varrho_s(0)$ and $P(A - A_s)^2 \varrho_s(0)$. After applying the operator P eq. (B.7) can be written as

$$(B.8) \quad b_s = \frac{(N-1)!}{N!} \left(\sum_{i \neq j \neq k \neq l} \theta_{ij} \theta_{kl} \right) \sum_{i \neq j \neq k \neq l} R_i^+ R_j^- R_k^+ R_l^- \varrho_s(0) + \\ + \frac{(N-3)!}{N!} \left(\sum_{i \neq j \neq k} \theta_{ij} \theta_{jk} \right) \sum_{i \neq j \neq k} R_i^+ [R_j^- R_k^+ + R_j^+ R_k^-] R_k^- \varrho_s(0) + \\ + \frac{(N-2)!}{N!} \sum_{i \neq j} \theta_{ij}^2 \sum_{i \neq j} R_i^+ R_i^- R_j^- R_j^+ \varrho_s(0).$$

By using the relations

$$\sum_{i \neq j} \theta_{ij} = 0, \quad \sum_j \Gamma_{ij} \sim N\alpha,$$

and expressing the sums of atomic operators in terms of collective operators, we obtain in limit of large N

$$(B.9) \quad b_s \sim \frac{1}{N^2} \left(\sum_{i \neq j} \theta_{ij}^2 \right) \left\{ \frac{2}{N^2} \left[R^+ R^- R^+ R^- - 2R^+ R \left(\frac{N}{2} + R_3 \right) + \right. \right. \\ \left. \left. + 2NR_3 - N \left(R^+ R^- - \frac{N}{2} + R_3 \right) \right] + \left[\frac{N}{2} \left(\frac{N}{2} + 1 \right) - R^2 \right] \right\} \varrho_s(0).$$

The quantity

$$\frac{1}{N^2} \sum_{i \neq j} \theta_{ij}^2 = \frac{1}{N^2} \sum_{i \neq j} [\Gamma_{ij}^2 - (1 - \beta)^2]$$

is zero in the two cases $\beta = 0$ and $\beta = 1$. In the general case, for large samples, we can make the approximation

$$\sum_{i \neq j} \Gamma_{ij}^2 = \sum_{i,j} \Gamma_{ij}^2,$$

then

$$(B.10) \quad \frac{1}{N^2} \sum_{i \neq j} \theta_{ij}^2 \simeq \beta(1 - \beta).$$

In these conditions, the first term in brackets of eq. (B.9) is seen to be of order $1/N$ against a_s . So, we have to compare the quantity

$$(B.11) \quad \beta(1 - \beta) \left[\frac{N}{2} \left(\frac{N}{2} + 1 \right) - R^2 \right] \varrho_s(0)$$

with a_s . It is easy to see that this quantity is always negligible against a_s . When the system has initially the maximum co-operation number $N/2$, eq. (B.11) is zero. When initially the co-operation number is minimum, *i.e.* for

$$R^2 \varrho_s(0) = \frac{3N}{4} \varrho_s(0),$$

then

$$a_s = \left[(1 - \beta) R^2 + \beta \frac{N}{2} \right]^2 \varrho_s(0) = \frac{N^2}{4} \left[\frac{3}{2} (1 - \beta) + \beta \right]^2 \varrho_s(0) \sim \frac{N^2}{4} \left[1 + \frac{b}{N} \right] \varrho_s(0),$$

whereas eq. (B.11) is

$$\beta(1 - \beta) \frac{N^2}{4} \simeq \frac{N^2}{4} \frac{b}{N} \varrho_s(0),$$

and then negligible against a_s .

APPENDIX C

We have to apply the symmetrization operator P on the operator $\sum_{i,j} \Gamma_{ij} R_i^+ R_j^-$ where Γ_{ij} is a function of positions of atoms i and j ; let

$$(C.1) \quad E = P \sum_{i,j} \Gamma_{ij} R_i^+ R_j^- = \frac{1}{N!} \sum_s S^{-1} \Gamma_{ij} R_i^+ R_j^- S,$$

where S is defined by eq. (2.5). From the relation (2.7b), E can be also written as

$$(C.2) \quad E = \sum_{i,j} \Gamma_{ij} \sum_{r,m,m',\eta} |\eta rm\rangle \langle \eta rm'| \sum_{\lambda} \frac{1}{n_r} \langle \lambda rm | R_i^+ R_j^- | \lambda rm' \rangle.$$

In order to calculate E , we will consider the expression (C.2). Any matrix element $\langle \lambda rm | R_i^+ R_j^- | \lambda rm' \rangle$ obeys the selection rule $\Delta m = 0$; for given (λ, r, m) it is generally dependent on the labels (i, j) and on the arbitrary choice of the orthonormal set $|\lambda rm\rangle$ ($\lambda = 1, \dots, n_r$). When the summation over λ is performed, $\sum_{\lambda} \langle \lambda rm | R_i^+ R_j^- | \lambda rm \rangle$ must be independent of the way in which the set $|\lambda rm\rangle$ has been constructed. For any S which is unitary in the representation realized by $\{|\lambda rm\rangle\}$, we have

$$(C.3) \quad \sum_{\lambda} \langle \lambda rm | R_i^+ R_j^- | \lambda rm \rangle = \sum_{\lambda} \langle \lambda rm | S^{-1} R_i^+ R_j^- S | \lambda rm \rangle.$$

By using the relation

$$(C.4) \quad R_j^{\pm} S = S R_i^{\pm},$$

where the label l can be different or not from the label j , depending on the permutation S , we show that $\sum_j \langle \lambda rm | R_i^+ R_i^- | \lambda rm \rangle$ and $\sum_j \langle \lambda rm | R_k^+ R_j^- | \lambda rm \rangle$ ($k \neq j$) are respectively independent of the label i and the labels (k, j) . We introduce this result into eq. (C.2) and we obtain

$$(C.5) \quad E = \left[\left(\sum_{i \neq j} \Gamma_{ij} \right) \sum_{r,m,\eta} |\eta rm\rangle \langle \eta rm| \sum_{\lambda} \frac{1}{n_r} \langle \lambda rm | R_k^+ R_l^- | \lambda rm \rangle + \right. \\ \left. + \left(\sum_i \Gamma_{ii} \right) \sum_{r,m,\eta} |\eta rm\rangle \langle \eta rm| \sum_{\lambda} \frac{1}{n_r} \langle \lambda rm | R_p^+ R_p^- | \lambda rm \rangle \right],$$

where p, k, l are arbitrary with the restriction $k \neq l$. Equation (C.5) shows that the symmetrization operator P factorizes the coefficients depending on atomic positions. This will be true when P is applied on any operator $\sum f(\{r_i\}) g(\{R_i\})$.

By using the well-known relations

$$(C.6) \quad \begin{cases} R^+ R^- |\lambda rm\rangle = (r+m)(r-m+1) |\lambda rm\rangle, \\ \sum_{i=1}^N R_i^+ R_i^- |\lambda rm\rangle = \left(\frac{N}{2} + m \right) |\lambda rm\rangle, \end{cases}$$

we easily obtain

$$(C.7) \quad \begin{cases} \sum_{\lambda} \langle \lambda rm | R_i^+ R_i^- | \lambda rm \rangle = \frac{n_r}{N} \left(\frac{N}{2} + m \right), \\ \sum_{\lambda} \langle \lambda rm | R_i^+ R_j^- | \lambda rm \rangle = \\ = \frac{n_r}{N(N-1)} \left[(r+m)(r-m+1) - \left(\frac{N}{2} + m \right) \right] \quad (i \neq j). \end{cases}$$

Introducing (C.7) and (C.8) in eq. (C.5) we finally obtain

$$(C.8) \quad E = \frac{1}{N(N-1)} \left(\sum_{i,j} \Gamma_{ij} - N \right) R^+ R^- + \frac{N}{N-1} \left(1 - \frac{1}{N^2} \sum_{i,j} \Gamma_{ij} \right) \sum_k \Gamma_{ij} R_k^+ R_k^-.$$

Equation (C.8) could be directly derived by applying the operators S on the spin variables in eq. (C.1). The above derivation shows that the permutations S only act on spin variables, leaving unchanged the atomic positions, *i.e.*

$$S^{-1} \Gamma_{ij}(r_i, r_j) R_i^+ R_j^- S = \Gamma_{ij}(r_i, r_j) S^{-1} R_i^- R_j^+ S.$$

● RIASSUNTO (*)

Si studia l'emissione spontanea di radiazione da parte di sistemi estesi di N atomi (o molecole) usando il metodo dell'equazione maestra. Per ogni atomo considerato come spin $\frac{1}{2}$ si coinvolgono due stati interni. Per manipolare l'equazione maestra si usano le proprietà delle rappresentazioni non riducibili del gruppo di permutazioni sulle variabili di spin. Adottando un'eccitazione uniforme per gli atomi si può definire una parte rilevante dell'operatore di densità atomica $\varrho(t)$. Questa parte rilevante di $\varrho(t)$ è invariante rispetto a ogni permutazione sulle variabili di spin e obbedisce a una facile equazione del moto che si dirà equazione maestra simmetrizzata, o S.M.E. Questa equazione è un'interpolazione delle equazioni maestre rispettivamente per un decadimento incoerente e un decadimento cooperativo in piccoli campioni. Si discute anche la preparazione del sistema atomico. La S.M.E. varrà per sistemi preparati sia con processi incoerenti di eccitazione, sia con intensi impulsi luminosi che lasciano il sistema in stati fortemente eccitati. Però il modello suddetto non si userà per atomi condotti a stati prossimi alla superradianza. Per grandi valori di N si danno soluzioni approssimate della S.M.E. Si trova che gli effetti di cooperazione appaiono per concentrazioni maggiori di quelle predette in lavori precedenti e il disaccordo è originato dal fatto che nella S.M.E. il numero atomico di cooperazione non è costante *a priori*. Per grandi concentrazioni il processo incoerente, ancora prevalente all'inizio dell'emissione, provoca un ritardo dell'emissione cooperativa. Nel decadimento atomico veloce si trova un andamento di sistemi descritti dalla S.M.E. identico a quello semi-classico. Si considerano anche effetti di allargamento non omogeneo, nel qual caso il processo incoerente smorza l'emissione cooperativa eccetto che alle altissime concentrazioni. Infine, si descrivono degli aspetti di rivelazione del campo radiativo per apparati rivelatori lontani dalla sorgente.

(*) *Traduzione a cura della Redazione.*

Подход, использующий симметризованное определяющее уравнение, к спонтанному излучению.

Резюме (*). — Исследуется спонтанное излучение из протяженных систем N атомов (или молекул), используя подход определяющего уравнения. Рассматриваются два внутренних состояния для каждого атома, который, считается, имеет спин $\frac{1}{2}$. Чтобы

(*) *Переведено редакцией.*

трактовать определяющее уравнение, мы используем свойства неприводимых представлений группы перестановок по спиновым переменным. Предполагая, что рассматриваемые атомы эквивалентно возбуждены, можно определить соответствующую часть оператора $\varrho(t)$ атомной плотности. Эта рассматриваемая часть $\varrho(t)$ является инвариантной относительно любой перестановки спиновых переменных. Этот оператор подчиняется уравнению движения, которое мы будем называть «симметризованным определяющим уравнением». Это уравнение представляет интерполяцию между определяющим уравнением для некогерентного распада и уравнением, относящимся к кооперативному распаду в малых образцах. Также обсуждается приготовление атомной системы. Симметризованное определяющее уравнение будет справедливо для атомных систем, приготовленных либо в результате процесса некогерентного возбуждения, либо интенсивными световыми импульсами, которые переводят систему в высокие возбужденные состояния. Таким образом, наша модель не будет использоваться, когда атомы приготовлены в сверхизлучающем состоянии. Приводятся приближенные решения симметризованного определяющего уравнения в пределе больших N . Получается, что кооперативные эффекты возникают для большой концентрации, чем предсказывалось в предыдущих работах. Это расхождение является следствием того факта, что в симметризованном определяющем уравнении число атомных коопреций не является априори постоянным. Для больших концентраций некогерентный процесс, который преобладает в начале излучения, приводит к запаздыванию кооперативного излучения. Получено, что в течение быстрого распада атомов поведение системы, описываемой симметризованным определяющим уравнением, оказывается тождественным квази-классическому. Также рассматриваются эффекты неоднородного уширения. В этом случае некогерентный процесс ослабляет кооперативное излучение, за исключением случая очень больших концентраций. В заключение описываются аспекты детектирования поля излучения системой детекторов далеко от источника.

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